Experimental Evidence of Critical Behavior in Cluster Fragmentation Using an Event-by-Event Data Analysis

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Using a recently developed multicoincidence technique, the hydrogen cluster fragmentation resulting from a collision between a high energy H_{25}^+ ion with a C_{60} target is investigated on an event-by-event basis. For a sample of 6000 collisions, statistical methods (conditional moments, scaled factorial moments) are applied to these data sets. The results obtained provide strong experimental evidence for the presence of a critical behavior in these finite systems. [S0031-9007(98)07568-1]

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Because of the general relevance in science, interest in the observation and characterization of critical behavior in finite systems, in particular, in nuclei and in clusters, has increased rapidly in the past years. Although small clusters do not exhibit exactly the first-order phase transition known in infinite matter, Kunz and Berry [1] have demonstrated that clusters may show a dynamic "solid-liquid" phase equilibrium which becomes a first-order phase transition for large cluster sizes. Furthermore, by using molecular dynamics simulations to study cluster fragmentation, the question on the existence and the nature of a finite-system analog of a second-order phase transition [2–5] (i.e., the correlation length becoming infinitely large at the critical point of a phase transition in normal macroscopic systems) has been addressed. Moreover, as such critical behavior is expected [6] to manifest itself similar to the nuclear case in the characteristics of the fragment mass distribution resulting from hot fragmenting clusters, several experimental studies have been performed on the fragmentation of clusters induced by collisions under different experimental conditions [7–10]. In all of these, a power law falloff as a function of fragment cluster size p, i.e., $p^{-\tau}$, has been observed with a critical exponent in close proximity to the critical exponent 2.6 found in nuclear fragmentation experiments [11–13] and with prediction ($\approx$2.23) from Fisher's droplet model [14]. Although these observations have been taken as a strong hint for the occurrence of critical behavior reminiscent of a second-order phase transition in an infinite system, the mass yield shape alone cannot be considered as conclusive proof [4]. There exist several further predictions concerning the outcome of collisions close to the critical point (for instance, the critical exponents, the intermittency signal, etc.), but these predictions can be verified only with data sets, where complete analysis of the fragments is available on an event-by-event basis. These extended analysis methods have been proved to be valuable tools in several theoretical cluster fragmentation studies [2,6,15], where the data have been generated by simulations. They have been already applied successfully to the analysis of nuclear collisions [16–19]. However, to date no experimental data from cluster collision experiments were available.

Using in the present study a recently developed multicoincidence technique for the measurement of the fragment size distribution induced by the collision of high energy H_{25}^+ ions with thermal C_{60} targets on an event-by-event basis, we have been able to obtain a first set of approximately 6000 completely analyzed collision events. This allows us to apply for the first time the method of conditional moments and of scaled factorial moments to measured cluster fragment distributions. We find ample experimental evidence for the presence of a critical behavior in these molecular systems.

The apparatus used here is shown in Fig. 1. Mass selected hydrogen cluster ions with an energy of 60 keV/amu are prepared in a high energy cluster ion beam facility consisting of a cryogenic cluster jet

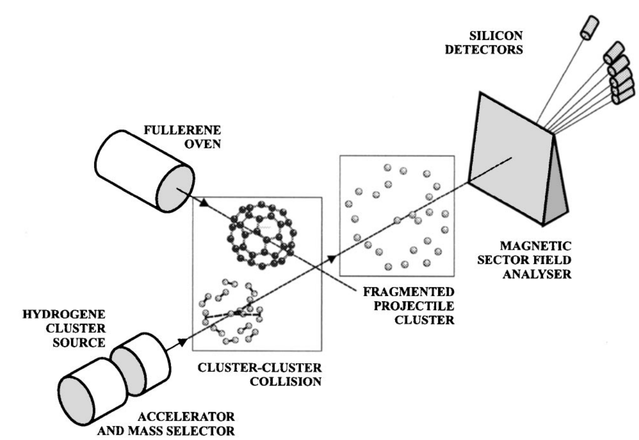

FIG. 1. A schematic view of the experimental setup.

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expansion source combined with a high performance electron ionizer and a two-step ion accelerator. After momentum analysis by a magnetic sector field, the mass selected high energy projectile beam consisting in the present study of H_{25}^{+} cluster ions is crossed perpendicularly by a C_{60} effusive target beam (see Fig. 1) produced by evaporation of pure C_{60} powder in a single-chamber molybdenum oven at about 675 °C. One meter behind this collision region the high energy hydrogen collision products (neutral and ionized) are passing a magnetic sector field analyzer approximately 0.3 μs after the collision event. The undissociated primary H_{25}^{+} cluster projectile ions or the neutral and charged fragments resulting from reactive collisions are then detected with a multidetector device consisting of an array of surface-barrier detectors located at different positions at the exit of the magnetic analyzer (see Fig. 1). This allows us to record simultaneously neutral and charged fragments detected in coincidence for each single collision event (for more experimental details, see Refs. [10] and [20]) irrespective of the nature of the collisional interaction, i.e., small or large impact parameters leading to rather "gentle" or "violent" collisions.

The data sets thus obtained ($\approx$6000 events) are then analyzed by first determining for each event the number of fragments $n(p)$ of size p and by constructing the conditional moments of the fragment size distribution as introduced by Campi [5],

$$M_i = \sum_p p^i n(p)\,, \tag{1}$$

where i is the order of the moment. The summation is over all the fragments in the event except the heaviest one. One important test to gain a first insight into the shape of the fragment size distribution and to indicate the extent of critical behavior is to then generate the relative variance γ_2 defined as

$$\gamma_2 = \frac{M_2 M_0}{M_1^2} \tag{2}$$

and to plot this quantity versus the temperature of the fragmenting system (the amount of energy in the fragmenting system). Since this quantity is not directly available, we are plotting here γ_2 versus the number of ions produced "NI" (see Fig. 2), as this number is indicative of the average energy in the excited cluster ion before fragmentation.

Figure 2 shows the present γ_2 data which resemble the earlier results reported by Campi [5] who considered the properties of a three-dimensional bond percolation system (cubic lattice with 125 sites). In fact, Campi [21] was the first to suggest that the methods developed in percolation theory may be of relevance to the analysis of multifragmentation data. In particular, it was shown that quantities that display, close to the critical point, divergent behavior in the macroscopic system (i.e., a phase transition of second order involving an infinitely large correlation length) are at least exhibiting a resonancelike behavior in finite size systems (with the corresponding correlation length being of the size of the system itself).

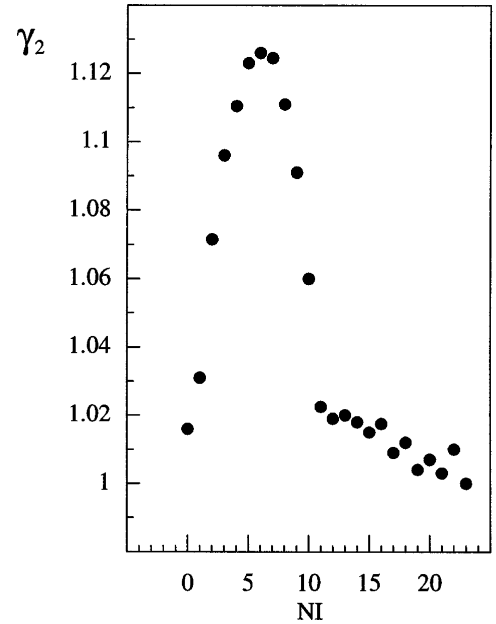

FIG. 2. The relative variance γ_2 versus the number of ions produced, NI, for the fragmentation of H_{25}^{+} hydrogen cluster induced by a collision with a C_{60} cluster at high velocity ($\approx c/88$).

In accordance with these considerations γ_2 has been demonstrated not only for percolation systems but also for simulated fragmentations of nuclei [18] and clusters [2] to have a peak around the critical point implying that the fluctuations are largest for critical conditions. The present experimental data are in good agreement with these predictions and results, and the occurrence of a peak in Fig. 2 at around NI $= 7$ confirms that at around this NI value the corresponding decaying hydrogen cluster ions are within the critical zone. At this point we can already conclude therefore that hot cluster ions are fragmenting similar to finite size systems which show a second-order phase transition for infinite size.

Because we are dealing in the present case with finite systems, additional methods are necessary to distinguish between dynamical fluctuations and statistical fluctuations. Thus in a next step we apply to our data sets one of the most convincing methods developed to analyze and categorize fluctuations and correlations, i.e., the analysis of event-by-event data in terms of intermittency. We now briefly outline the procedure. Intermittency is a statistical concept and it corresponds to the existence of nonstatistical fluctuations which have self-similarity over a broad range of scales. The relevant information can be deduced from the horizontally scaled factorial moments (HSFM) which measure the properties of dynamical fluctuations without the bias of statistical fluctuations (deviations of the fluctuations from a Poissonian distribution) [17,22] given by

$$(\mathrm{HSFM})_i = M^{i-1} \sum_{k=1}^{M} \frac{n_k(n_k - 1)\cdots(n_k - i + 1)}{N(N - 1)\cdots(N - i + 1)}, \quad (3)$$

where

$$M = \left\{\frac{X_{\max}}{\delta s}\right\} + 1. \quad (4)$$

$X_{\max}$ is an upper value of a characteristic quantity of the system (e.g., total mass, maximum transverse energy, momentum, velocity, etc), and i is the order of the moment. The total interval $0-X_{\max}$ ($1-p_{\max}$ in case of size distributions) is divided into $M = [(X_{\max}/\delta s) + 1]$ bins of size δs, whereby n_k is the number of particles in the kth bin of an event. In Eq. (3), N is the total number of fragments in the event.

If self-similar fluctuations exist at all scales δs, the scaled factorial moments follow the power law

$$(\mathrm{HSFM})_i \propto \delta s^{-\lambda_i}, \quad (5)$$

where λ_i are called intermittency exponents. Thus, the intermittent behavior (intermittency signal) is defined as a linear rise in a plot of $\ln(\mathrm{HSFM})_i$ versus $-\ln(\delta s)$.

In Fig. 3 we plot the scaled factorials moments ln(HSFM) versus $-\ln(\delta s)$ for the values of the resolution δs from 1 to 24. For events from outside of the critical region (NI $>$ 11), the logarithms of the $(\mathrm{HSFM})_i$ are independent of δs and therefore $\lambda_i = 0$. This corresponds to a situation with no intermittency signal and the fluctuations present are only statistical.

Conversely, the situation is clearly different for events belonging to the critical region determined by the resonance in Fig. 2, i.e., $2 \leq \mathrm{NI} \leq 11$. In this case, the logarithms of the $(\mathrm{HSFM})_i$ are increasing for each order i linearly versus $-\ln(\delta s)$ thus indicating the presence of an intermittency signal. Before discussing the results given in Fig. 3(b) in more detail, we would like to make the following comments. It turns out that the $\ln(\mathrm{HSFM})_i$ values for $\delta s = 1$ and 2 deviate from the linear behavior in the log-log pot [values for $\delta s = 1$, larger than 0.2, are not plotted in Fig. 3(b)]. Such a deviation has also been observed in similar plots obtained for experimental and simulated data on fragmentation of gold nuclei [16,17]. According to DeAngelis *et al.* [17] this deviation is presumably due to the fact that small δs correspond to the limit of the resolution, and, therefore, these data points are not included in the derivation of λ_i. Besides the results on nuclei fragmentation, such a linear dependence of the $\ln(\mathrm{HSFM})_i$ versus $-\ln(\delta s)$ has been obtained by Kondratyev and Lutz [2] in a theoretical study about fragmentation of argon clusters of a size of 100. It is interesting to note that based on experimental data a similar behavior is observed here with a cluster size of only 25. This demonstrates the usefulness of this approach even for rather small systems.

Moreover, for simulating nuclear fragmentation, Belkacem *et al.* [18] reported plots of $\ln(\mathrm{HSFM})_i$ versus $-\ln(\delta s)$ for different times after the primary excitation event. The values of the $(\mathrm{HSFM})_i$ are shown to decrease with the increase of time between the collision and the detection of the fragments, but the intermittency exponents are shown to remain about constant after some time (see Fig. 16 in Ref. [18]). It is noteworthy that the present results [Fig. 3(b)] are very similar to those obtained by Belkacem *et al.* [18] at late times, i.e., concerning the ordering and the slope as a function of i.

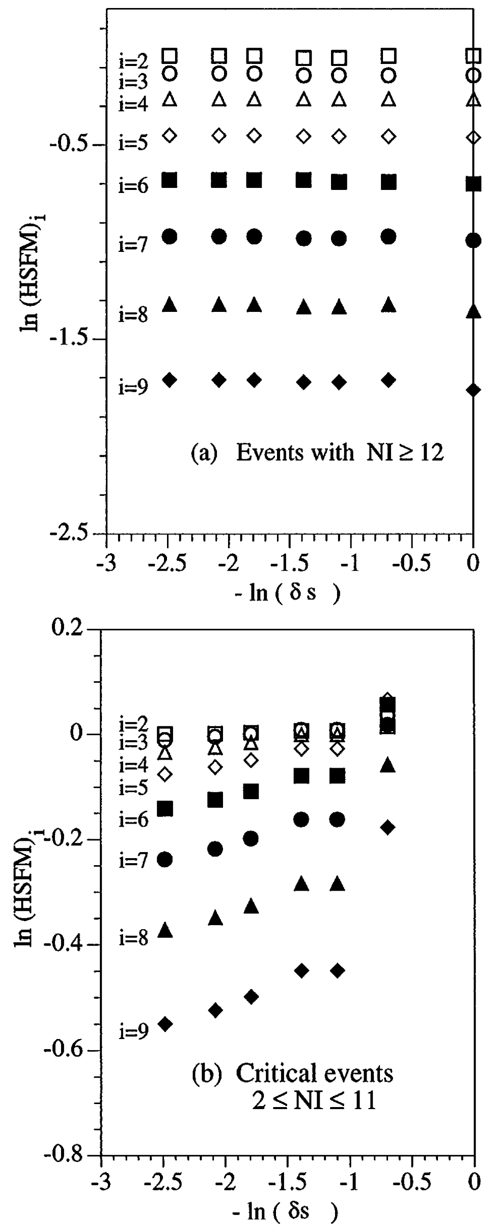

FIG. 3. Log-log plot of the horizontally scaled factorial moments $(\mathrm{HSFM})_i$ versus the resolution δs (see text). The $(\mathrm{HSFM})_i$ are calculated for (a) events from outside of the critical region NI $>$ 11, and for (b) events belonging to the critical region as determined by the resonance in Fig. 2, i.e., $2 \leq \mathrm{NI} \leq 11$.

Finally, an important quantity related to the intermittency exponent λ_i is the anomalous fractal dimension [4,17,18] defined as

$$d_i = \frac{\lambda_i}{i - 1}. \quad (6)$$

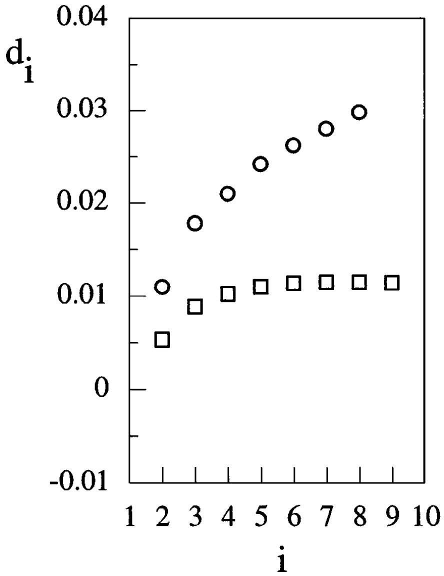

FIG. 4. The anomalous fractal dimensions d_i versus the order of the moment i deduced from two different experimental data sets. Open squares correspond to the hydrogen cluster fragmentation (this work). Open circles correspond to gold nuclei fragmentation (Ref. [16]).

As shown by Belkacem *et al.* [18], this quantity is insensitive to secondary reactions (evaporation from excited fragments) as discussed above, where we have shown that the λ_i are constant for longer times. Nevertheless, for different fragmentation mechanisms (monofractal or multifractal) the anomalous fractal dimension can exhibit different behaviors with i, i.e., d_i independent of i corresponding to a monofractal, second-order transition in the Ising model, and d_i proportional to i corresponding to multifractal, cascading processes [4]. Hence, analysis of the intermittency exponents in terms of d_i will yield interesting information on the decay mechanism of the excited system. In Fig. 4, the anomalous fractal dimensions are plotted versus i for the present cluster data. For comparison we include data obtained for gold nuclei fragmentation from Ref. [16]. In both cases no unambiguous classification according to the above mentioned dependencies of d_i on i can be deduced. The present anomalous fractal dimension is observed to increase and then to saturate with increasing i. A similar behavior is also observed for gold nuclei fragmentation, but the saturation effect is more pronounced in the cluster case. This effect has been related, in the nuclei fragmentation case, to finite size effects [17,18].

In conclusion, this first analysis of cluster fragmentation based on fully determined event-by-event data sets shows that the power law observed in the fragment size distribution of the finite system studied here is connected to a critical behavior reminiscent to a second-order phase transition in an infinite system. Both the Campi conditional moments analysis and the scaled factorial moments analysis lead to this conclusion. In addition, close similarity in the fragmentation behavior of clusters and nuclei is not only observed in the inclusive fragment size distribution, but also in the event-by-event analysis. Moreover, the application of these characterizations of the fragment size distribution—as demonstrated here for the cluster case—appears to be successful for quite different microscopic systems (at different scales) and thus also applicable to the fragmentation of macroscopic objects (see the observation of a power law behavior in the fragmentation of platelike objects [23]). Finally, the success of the statistical approach for the experimental characterization of a critical behavior in a finite system should be a stimulus to improve the understanding of the dynamics of complex systems, when the correlation length between the constituents is of the size of the system itself.

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